In this thesis, interfacial modification is used to improve the performance of p-i-n-structured lead-based perovskite solar cells. Organic-inorganic halide perovskites have been shown as highly-efficient light harvesting materials. Many reports are available, illustrating their outstanding optoelectronic properties. However, the performance of multilayered thin-film devices using perovskites is dependent on interfacial properties within the device, determining film formation, charge transport and charge extraction.

In this work, device interfaces are modified in order to improve the efficiency of lead-based perovskite solar cells. Current density-voltage (JV) characterization is used to investigate changes of the device characteristics. Major improvements are observed by the adjustment of the interfacial energetics between the electron transport layer (ETL) and the metal cathode. By inserting a thin interlayer of the low work function metal calcium, the energetic barrier for electron extraction is reduced. Consequently, large improvements in fill factor are observed based on the prevention of S-shaped JV curves. Similar observations are made by the introduction of organic cathode buffer layers of PEIE or PFN-P1 and the hole blocking layers of BCP and BPhen.

For the first time a series of polymeric cross-linkable hole transport materials (x-HTM) is introduced. The advantage of the tuneability of their highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) by exchanging molecular substituents is applied to study their hole extraction and electron blocking abilities. It is shown, that efficient hole extraction depends on the energy level difference ΔE_{HT} between the HOMO level of the HTM and the valence band of the $MAPbI_3$ perovskite. By using QUPD the best hole extraction is obtained in the investigated series. All other tested x-HTMs with lower HOMO levels showed reduced hole extraction, indicated by S-shaped JV curves. Introducing TAPC derivatives with higher LUMO levels leads to improved electron blocking at the hole transport layer (HTL) interface indicated by an increase in V_{OC} and $J_{SC.}$

Nevertheless, the application of a 5 nm thin layer of QUPD leads to high-efficient perovskite solar cells with a shown record power conversion efficiency (PCE) up of 17.3% using the $MAPbI_3$ perovskite absorber.

By application of the multiple-cation perovskite of $(CsPbI_3)_5[(FAPbI_3)_{83}(MAPbBr_3)_{17}]_{95}$ in combination with QUPD even 19.2% PCE is shown. Thus, the competitiveness of the x-HTL materials compared to commercially available materials like poly-TPD or PTAA is highlighted.

JV results are shown to be affected by device pre-treatments by light or voltage and thus causing changes in the charge distribution within the device. Thus, the development of a maximum power point tracking (MPPT) algorithm is presented, in order to obtain steady-state solar cell characteristics. Investigations about the irradiation intensity-dependent solar cell characteristics, as well as examination of the definition of the illumination area by the application of shadow

masks, reveal important aspects in avoiding mischaracterization in JV measurements of p-i-n-structured perovskite solar cells.